

# Elastic properties and equation of state for polycarbonate by high-pressure Brillouin spectroscopy



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## ABSTRACT

Although the conventional vibrational spectroscopic techniques of Raman and infrared spectroscopy are generally used to analyze the material properties of polymers, the physical and mechanical information that can be obtained through these techniques is very scarce. Since elastic information and equation of state are important for the application of polymers, we carried out Brillouin spectroscopy coupled with a diamond anvil cell. The pressure dependence of elastic properties for an amorphous polycarbonate with and without pressure medium was investigated for pressure up to 11 GPa. A substantial collapse of free volume under the anisotropic pressure was ascertained with the different slopes of elastic constants, Young's moduli, and bulk moduli at low and high pressures. From the Brillouin frequency shifts, we extracted  $C_{11}$  and  $C_{12}$  elastic constants, Young's moduli, shear moduli, bulk moduli, Poisson's ratios, and their pressure dependences.  $P$ - $V$  isotherms were then constructed and fitted to two semi-empirical equations of state to extract the isothermal bulk modulus and its pressure derivative.

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## 1. Introduction

These days many kinds of polymers have been applied to various fields and heavily used in daily life. Among the polymers, polycarbonate (PC) is often viewed as the typical engineering thermoplastic due to its advantages, such as combination of toughness, high strength, high heat-deflection temperature, and transparency, in application. Moreover, since it has also reasonably good temperature resistance and dimensional stability, it has been commercialized several products such as glazing, safety shields, safety goggles, cockpit canopy, lenses, casings and housings, light fittings, CDs, and DVDs [1,2]. However, PC has also disadvantage in applications owing to poor fatigue, wear and tear, and is prone to environmental stress cracking. These facts indicate that it is necessary to understand its mechanical property exactly, i.e. elastic property and equation of state (EOS) for further wide applications. In particular, the EOS is the most fundamental equation in providing a basis for the accurate determination of physical properties of any material.

Structural and mechanical properties of crystalline materials

with long-range order under extreme condition (high pressure or/and temperature) have been actively studied in physics, chemistry, material science, geosciences, and planetary science using x-ray diffraction (XRD) combined with a diamond anvil cell (DAC) [3–5]. However, for amorphous materials such as polymers with short-range order, XRD is not applicable to determine the EOS, because the distinct diffraction peaks from interference effect don't appear. Alternatively, although a dilatometer has been generally utilized to carry out the  $P$ - $V$ - $T$  measurement of polymers, its maximum working pressure is limited to 0.2 GPa which is much lower compared with that generated by DAC [6].

Brillouin spectroscopy coupled with a DAC has been applied to investigate the elastic properties and to construct the EOS of Kel-F 800 polymer up to the pressure of 85 GPa [7,8]. However, experimental  $P$ - $V$  EOS data and elastic property of PC for the pressure over 0.2 GPa is not reported yet to our knowledge. In order to study the  $P$ - $V$  EOS and elastic property of PC, we have carried out high-pressure Brillouin spectroscopy measurement with a DAC to determine two independent elastic constants and  $P$ - $V$  EOS. In particular, we report the new experimental method which enhances the accuracy of EOS significantly by measuring the scattering angle of laser by the sample inside the gasket hole of a DAC using the two Brillouin scattering geometries, i.e. forward symmetric and back scatterings, with a liquid standard sample. This

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new method has not been reported to and used by the Brillouin spectroscopy community up to now.

Furthermore, although it has been generally taken for granted that the pressure measurement at several positions of the gasket hole loaded with soft materials is good enough to guarantee the hydrostaticity of them, a generalized Cauchy relation is used to check the validity of omnidirectional stress for soft substances like PC [9].

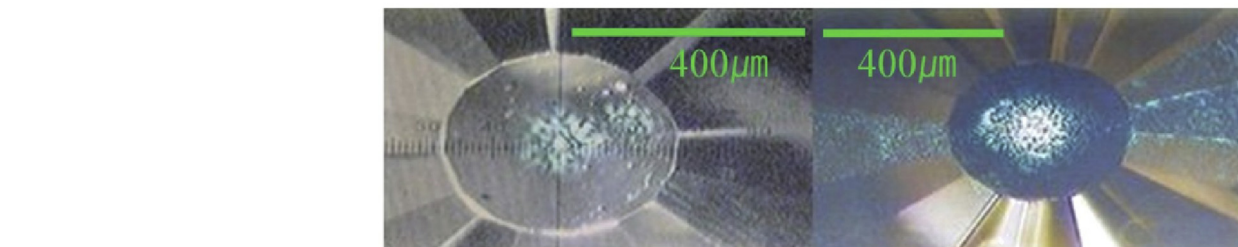
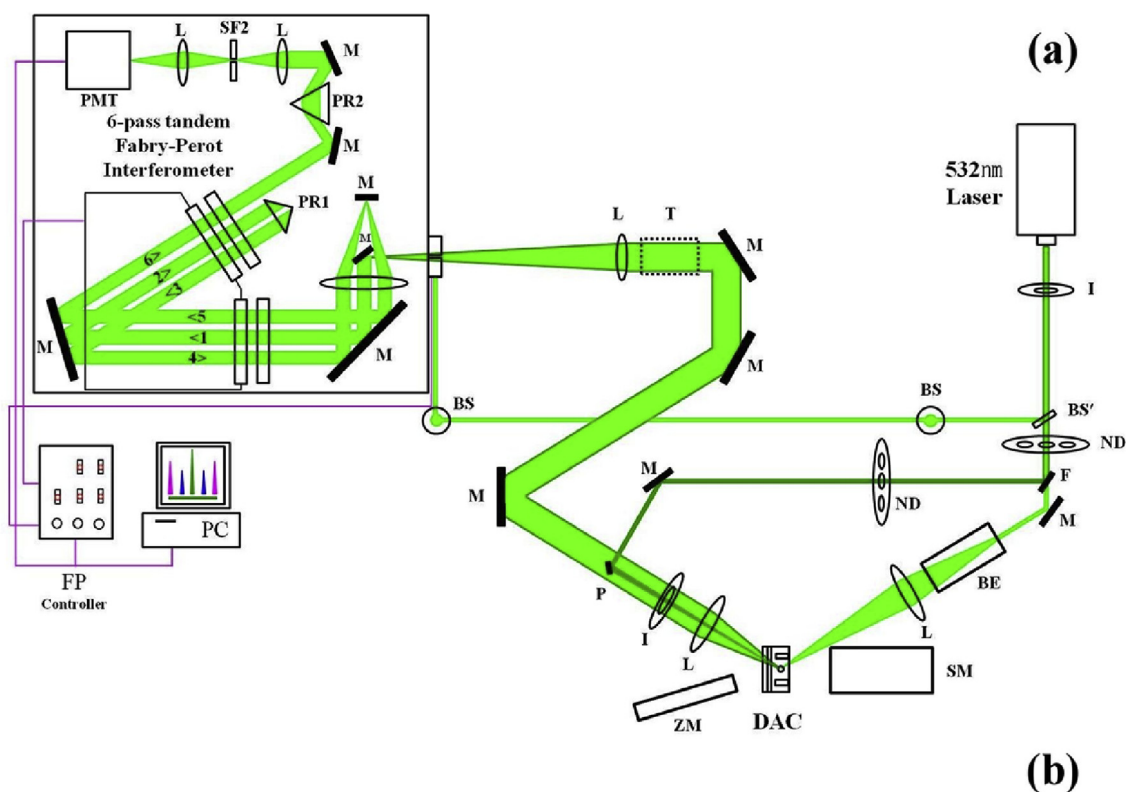
## 2. Experiments

The PC sample (PC CT301350) was purchased from Goodfellow Cambridge Limited and used without any further treatment or purification. There is no crystallinity in this sample.

A conventional tandem six-pass Fabry-Perot interferometer (TFP-1, JRS scientific instruments) was used to measure the Brillouin spectra; a diode-pumped solid-state laser (SLIM, Oxxius) operating with a wavelength ( $\lambda$ ) of 532 nm and power of 300 mW was used as an excitation source [5]. A micro-Brillouin scattering technique was adopted for forward symmetric scattering and

backward scattering geometries. The free spectrum range was adjusted to cover a wide frequency range. Fig. 1(a) shows the experimental setup for the high-pressure Brillouin spectroscopy and Fig. 1(b) shows the diameters of laser beam focused on the culet of diamond anvils contacting with each other. For the back scattering geometry, a single achromatic lens ( $\varnothing 50$  mm) focuses the laser beam to  $\sim \varnothing 100$   $\mu\text{m}$  and simultaneously collects the scattered lights by the sample. For the forward symmetric scattering geometry, an f-theta scanning lens coupled with a 10X beam expander (Edmund optics) is used to focus the laser beam to  $\sim \varnothing 17$   $\mu\text{m}$ . Since the laser beam is focused to tens of micrometer, the sample or any material scattering the focused laser can be heated. Therefore, a rotating filter wheel with neutral density filters is used to prevent the sample or ethyl alcohol in the gasket hole from being heated.

Ethyl alcohol of anhydrous ( $\leq 0.003\%$  water) from Sigma–Aldrich is used as a standard calibration sample for the determination of scattering angle. Fig. 2 shows the Brillouin spectra of the ethyl alcohol in the gasket hole of a DAC at room temperature (293 K) and ambient pressure. The determined scattering angle from this measurement is  $\theta = 62^\circ \pm 0.0^\circ$ . The detailed explanation



**Fig. 1.** (a) Experimental setup for high-pressure Brillouin spectroscopy. SF: spatial filter, PR: prism, M: mirror, L: lens, I: iris diaphragm, BS: beam steerer, BS': beam sampler, SM: stereo microscope, P: right angle prism, F: flipper, ND: neutral density filter, BE: beam expander, ZM: zoom microscope, T: telescope, and DAC: diamond anvil cell. Green for forward symmetric scattering and dark green for back scattering (b) Diameters of laser beam focused on the culet of diamond anvils contacting with each other for forward symmetric scattering (left) and back scattering (right). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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